#4

Piezoelectric Composites with High Sensitivity and High Capacitance for Use at High Pressures

Q. C. Xu, S. Yoshikawa, J. R. Belsick, and R. E. Newnham, *Member, IEEE*

Abstract—**A new type of piezoelectric composite has been developed for oceanographic applications. The composites have a large figure of merit ($d_h \cdot g_h$ or $d_h \cdot g_h/\tan\delta$), a large dielectric constant (K) and low dielectric loss, as well as great mechanical strength. A shallow cavity between the PZT ceramics and thick metallic electrode is designed to convert a portion of the z-direction stress into a large radial and tangential stress of opposite sign, thereby causing the d_{33} and d_{31} contributions to d_h to add rather than subtract, and raising the figure of merit. Theoretical stress analysis was carried out using an axisymmetric finite element method. Experimental results show that the $d_h \cdot g_h$, K and withstandable pressure are extremely high.**

I. Introduction

FOR many hydrophone applications, there is a great demand for piezoelectric composites with a high hydrostatic piezoelectric charge coefficient (d_h), high hydrostatic piezoelectric voltage coefficient (g_h), and high dielectric constant (K) as well as a high pressure tolerance. In the last decade, several piezoelectric ceramic–polymer composites with different connectivity patterns have been developed for hydrophone and medical transducer applications [1]–[4]. The advantages of these composites over ceramics include higher figure of merit $d_h \cdot g_h$ to enhance the sensitivity, increased mechanical compliance, smaller acoustic impedance for matching to water or tissue, and lower transverse electromechanical coupling coefficient to reduce cross-talk noise and improve directivity of the transducer array. Disadvantages of these ceramic-polymer composite transducers however, are lower dielectric constant and lower pressure tolerance than their ceramic counterparts.

Flextensional transducers composed of a piezoelectric ceramic and a shell structure exhibit good electro-acoustic performance [5], [6] in which the extensional vibration mode of a piezoelectric ceramic is coupled to the flexural vibration mode of a metal or polymer shell. The shell is used as a mechanical transformer for transforming the high acoustic impedance of the ceramic to the low acoustic impedance of the medium and for producing large volume velocity. Or, when operated in the reverse direction, the large velocity in the medium produces a high stress in the ceramic. All five types of flextensional transducers described in [5] and [6] are designed to operate in the low frequency range below 10 kHz.

Manuscript received August 15, 1990; revised February 11, 1991; accepted May 7, 1991. This work was supported in part by the Office of Naval Research and in part by the Center for the Engineering of Electronic and Acoustic Materials at the Pennsylvania State University.

The authors are with the Materials Research Laboratory, Pennsylvania State University, University Park, PA 16802.

IEEE Log Number 9102673.

TABLE I
COMPARISON OF $g_h \cdot d_h$ PRODUCT AMONG VARIOUS PIEZOELECTRIC MATERIALS

Material	$d_h \cdot g_h$ ($\times 10^{-15}$ m^2/N)
PZT-brass composite	~50,000
PZT	~110
$PbTiO_3$	~1,800
Voided Thick PVDF	~5,000

This paper describes a new type of piezoelectric ceramic-metal composite based on the principle of a flextensional transducer. The basic structure is described in Section II and has some similarity to a class V flextensional transducer [7]. A computer analysis of the stress in the composite was performed using the finite element method (FEM). The stress contours are described in Section III. In Section IV, the experimental results are presented to show that this type of composite can provide very high $d_h \cdot g_h$ or $d_h \cdot g_h/\tan\delta$, [8] together with a large capacitance and high withstanding pressure. Table I compares the $d_h \cdot g_h$ values of the PZT-metal composite with other commonly used transducer materials.

II. Basic Principle

As is well known, PZT ceramics have high d_{33} and d_{31}, but their d_h ($= d_{33} + 2d_{31}$) values are only about 45 pC/N because d_{31} and d_{33} have opposite signs. To enhance d_h, we have developed a PZT-metal composite with very shallow cavities between the PZT ceramic and thick metal electrodes that convert a portion of the z-direction stress into large radial stresses of opposite signs, thereby causing the d_{33} and d_{31} contributions to d_h to add rather than subtract, leading to high d_h.

A cross section view of the disk shaped PZT-metal composite is shown in Fig. 1. The cylindrically symmetric structure is designed to obtain an extensional vibration mode of PZT, and high hydrostatic pressure tolerance. The height of the shallow cavity h is less than 150 μm. The shallow cavity allows deformation of the metal electrode toward the ceramic disk by closing the cavity that reduces stress amplification in the PZT and prevents

0885-3010/91$01.00 © 1991 IEEE

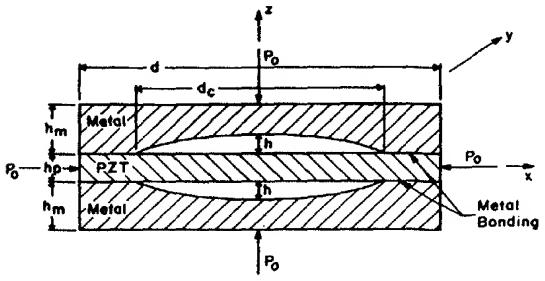

Fig. 1. Geometry of the composite.

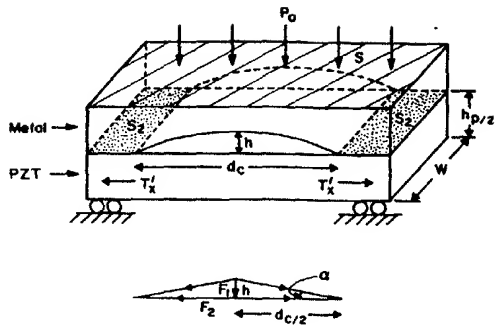

Fig. 2. Simplified model for calculating stress transformation.

breakdown during shockwaves or very high hydrostatic pressure. The concept of the stress transformation can be explained in a simple manner using the cross section in Fig. 2 and the following equations:

$$\frac{F_1}{F_2} = \frac{h}{\frac{d_c}{2}} = \frac{2h}{d_c} \tag{1}$$

$$F_1 = W \cdot \frac{d_c}{2} P_o, \quad F_2 = W \cdot \frac{h_p}{2} T'_x \tag{2}$$

$$T'_x = \frac{d_c}{2h} \cdot \frac{d_c}{h_p} P_o = N \frac{d_c}{h_p} P_o \tag{3}$$

$$N = \frac{d_c}{2h} \cong \frac{1}{\tan \alpha}. \tag{4}$$

Similarly

$$T'_y = N \frac{d_c}{h_p} P_o. \tag{5}$$

The extensional stress is considered as "negative" here. Let T_x be the x-direction stress in PZT, then

$$T_x \cong P_o - N \frac{d_c}{h_p} P_o \cong T_y \tag{6}$$

where P_o is the acoustic pressure and N is a stress amplification parameter; N is approximately equal to $1/\tan \alpha$, where α is the shallow cavity angle shown in Fig. 2; T_z is the z-component of stress in PZT and is given by

$$T_z = P_o. \tag{7}$$

The resulting polarization is $d_{31} T_x + d_{31} T_y + d_{33} T_z S_2/S$, where S is the surface area of the PZT and S_2 is the surface area of the metal-PZT bond.

Therefore

$$(d_h)_{\max} \cong d_{33} \frac{S_2}{S} + 2d_{31}\left[1 - \frac{Nd_c}{h_p}\right]. \tag{8}$$

This estimate of $(d_h)_{\max}$ explains the basic principle of the composite, but the experimental result of d_h is much smaller than $(d_h)_{\max}$, partly because N is much less than $1/\tan \alpha$ for a thick metal electrode.

The basic idea of the composite is to attempt to use both the d_{33} and d_{31} coefficients to obtain high d_h. Thick metal plates are used as a mechanical transformer to transform the stress direction and amplitude, and also to adjust the acoustic value from a low impedance in the z-direction to a high impedance in planar direction.

The lowest vibration mode of the composite is a flextensional mode determined mainly by the stiffness of the PZT in a planar mode and the equivalent mass of the metal plate. This equivalent mass is much larger than the real mass of the metal plate, because the vibration velocity at the central portion of the metal is much larger than the reference velocity in the PZT. The operating frequency range of the composite is dependent on this flextensional mode that is controlled by the cavity diameter d_c, the height of the cavity h, the thickness of the metal h_m and the stiffness of the ceramic in planar mode.

Since the withstanding hydrostatic pressure P_m is another important parameter for the underseas application, the stress amplification coefficient N cannot be designed too high. Transducers with large cavity diameters (d_c) have low flextensional resonant frequency, low P_m and high d_h.

The capacitance of the composite can be changed by adjusting the electrode area on the PZT surfaces, especially if the stress in the center portion of the PZT disk is small.

III. Stress Analysis with FEM

A theoretical analysis of the PZT-brass composite was performed using the finite element analysis program, AN SYS version 4.3 [9], [10]. A one-quarter axially-symmetric model is shown in Fig. 3. The mesh contained 640 quadrilateral-shaped elements with 729 nodal points. Half of them are in the PZT. The triangular points are used to "pin" the object and allow only parallel motion on the boundary when stresses are employed.

To simplify the analysis, the metal bonding layer is neglected, and a hydrostatic reference pressure of $P_o = 1$ is applied to the model. Fig. 4 shows the stress contours in the radial (R) direction with a quadrupole pattern in the brass, and a stress concentration factor of about 20 at the tip point of the PZT and the brass. In the PZT, there are only extensional stresses in the radial R direction and very small stresses in the central portion of the ceramic. The

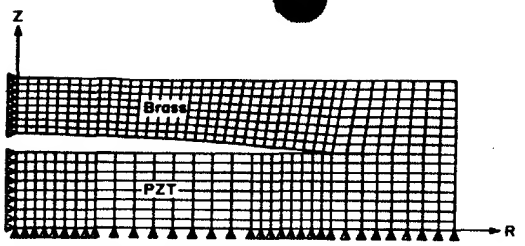

Fig. 3. Mesh used for modeling PZT-brass composites.

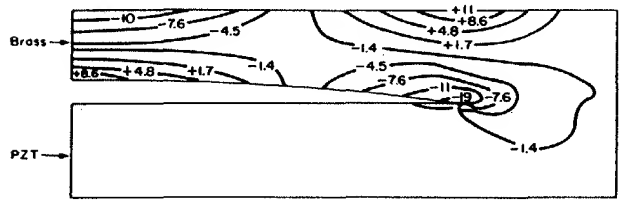

Fig. 4. Finite-element model of stress in R direction.

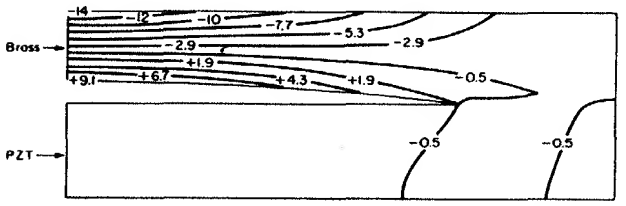

Fig. 5. Finite-element model of stress in ϕ direction.

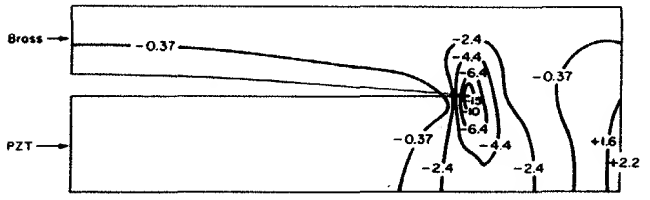

Fig. 6. Finite-element model of stress in z direction.

tangential stresses (ϕ direction) in Fig. 5 shows that there are bending stresses in the brass, and extensional stresses in the PZT. The stress contours in the z-direction shown in Fig. 6 indicate that there are undesirable extensional stresses in the PZT, and the stresses are concentrated with a factor of about 15 at the tip of the cavity between the PZT and brass. The stress analysis, which neglected the PZT-brass interface layer, shows that the material used as to bond the PZT and the brass should have greater compliance than brass or PZT in order to reduce the stress concentration factor and to obtain compressive stresses in the PZT along the z-direction, thereby forming large extensional stresses in the PZT along the R- and ϕ-directions. A thick bonding layer of a metal with lower elastic moduli metal leads to higher sensitivity in hydrophone applications.

IV. Experiments and Results

PZT-brass composite samples with dimensions (Fig. 1): d = 11 mm, h_m = 1.2 mm, h_p = 1.1 mm, h = 100–150 μm, and four different cavity diameters d_c = 7.6 mm (large cavity), 5.8 mm (middle cavity), 4.1 mm (small cavity), 2.5 mm (very small cavity) were fabricated for the experiments. In order to obtain a thick bonding layer, the brass plates were bonded to a PZT-5 disk with the capacitor electrode silver paste and fired at 600°C for 10 min. Brass was chosen for its lower thermal expansion coefficient (approximately 15 ppm/ °C). After cooling, the composite was encapsulated around its circumference with Spurrs epoxy resin and cured at 90°C for more than 8 hours. The composite was poled in oil at 150°C with a 2.5 kV/mm electric field for about 15 min.

The g_h coefficient was determined using a dynamic ac technique. An electromagnetic driver was used as an ac stress generator to apply pressure waves to the sample and a PZT standard, which were kept under a static pressure (up to 1000 psi (7 MPa)) with a hydraulic press. The charge produced by the sample and the standard were buffered with an impedance converter, and the resulting voltages measured on a Hewlett-Packard 3585A Spectrum Analyzer. The ratio of the voltages is proportional to the g_h coefficients. Accounting for the geometries of the sample and PZT standard, and the stray capacitance of the holders, the g_h coefficient of the sample was calculated. Using the measured values of g_h, the hydrostatic piezoelectric coefficient, d_h, was calculated from the relation, $d_h = \epsilon_0 K g_h$.

A question arises in how to compare the output of this transducer with other piezoelectrics. Since g_h is obtained from the output voltage and the thickness of the composite and standard samples, we have chosen the thickness of composite sample to be the same as the thickness of PZT disk in order to retain the same dielectric constant as PZT. Otherwise, g_h will be three times less and the apparent dielectric constant will be three times higher.

The experimental results presented in Figs. 7 and 8 show that large cavity sizes lead to very large d_h and g_h values. Moreover, the dielectric constant exceeded 1500 and tan δ was less than 0.025. Fig. 9 shows the relationship between the d_{33} value, measured at center point of the sample with a Berlincourt d_{33} meter using the electromagnetic driver operating at a frequency of 100 Hz, and the d_h value measured by the method just described. The d_{33} value increases markedly with cavity diameter. Fig. 10 shows that the frequency of the lowest flextensional mode decreases as the cavity diameter increases. Therefore, the larger cavity diameter composites possess a lower operating range.

Because the thermal expansion coefficient of brass is larger than the PZT (approximately 5-7 ppm/ °C), compressive prestresses are applied to PZT during the bonding process in R and ϕ directions perpendicular to the pol-

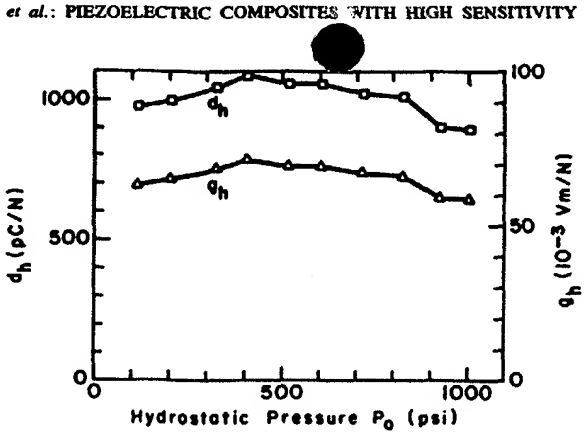

Fig. 7. Hydrostatic pressure dependence of d_h and g_h. Large cavity, d_c = 7.6 mm, K = 1700.

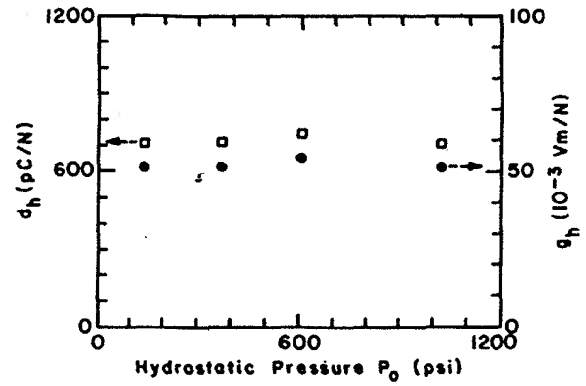

Fig. 8. Hydrostatic pressure dependence of d_h and g_h. Small cavity, d_c = 4.1 mm, K = 1560.

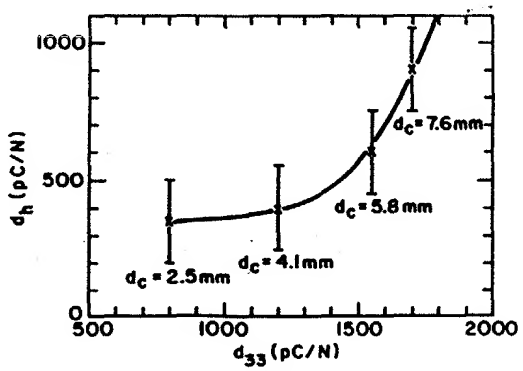

Fig. 9. The d_h dependence of d_{33} at center point of the disk.

ing direction. These prestresses help to maintain the polarization in the PZT. Fig. 11 shows that aging under hydrostatic pressure at 350 psi (about 2.5 MPa) was very small.

Lastly, a planar array was made for testing by embedding four composite samples with large cavities in epoxy resin (Eccogel 1365-25, Emerson and Cummings, Inc.) (Fig. 12). The admittance and conductance of the array

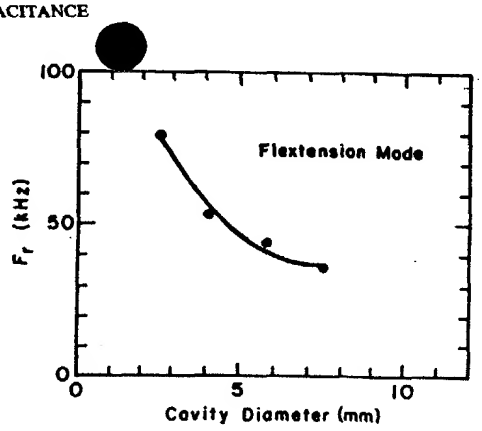

Fig. 10. First flextensional frequency dependence of cavity diameter.

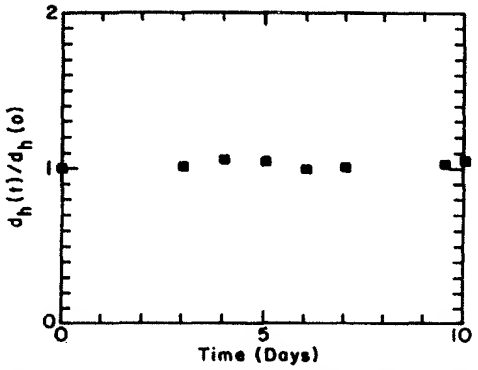

Fig. 11. Aging under hydrostatic pressure. Hydrostatic pressure P_m = 350 psi.

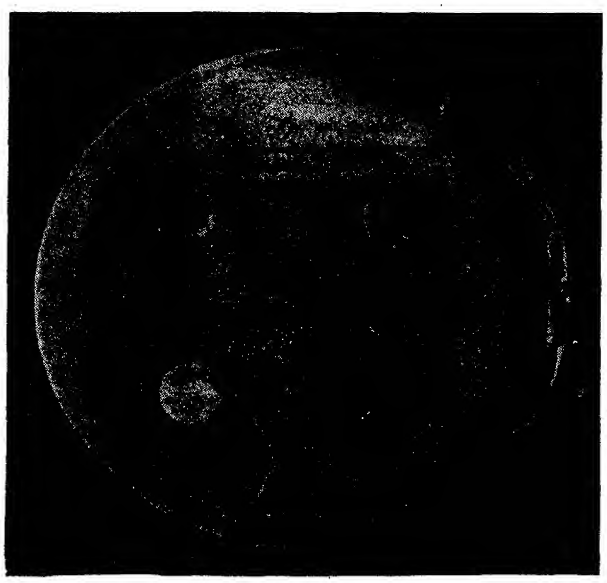

Fig. 12. Flexible array incorporating four composite transducers.

in air and in water presented in Figs. 13 and 14 show that the lowest flextensional mode was higher than 30 kHz and that the resonant peak in water was flat. Since conduct-

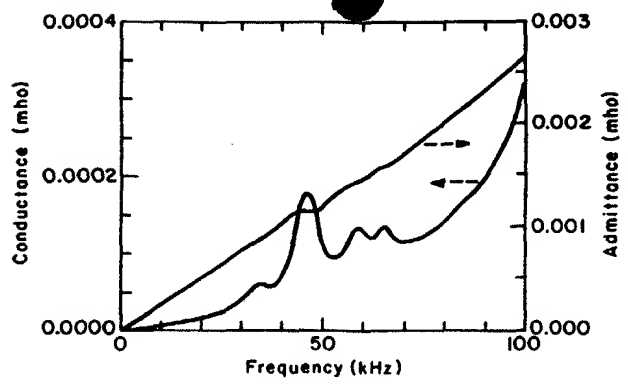

Fig. 13. Conductance and admittance of the array in air. Four elements array in air.

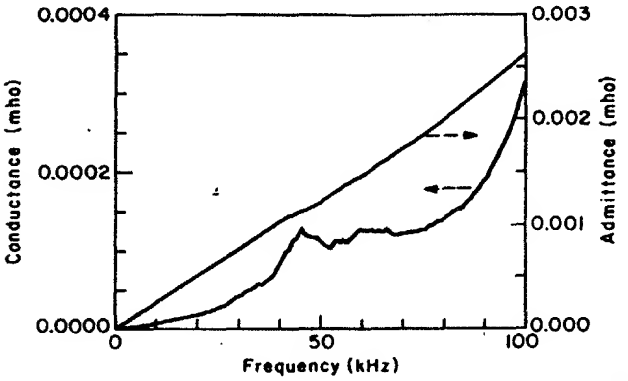

Fig. 14. Conductance and admittance of the array in water. Four elements array in water.

ance exhibit only a weak resonance peak, a flat receiving response is expected to extend to more than 20 kHz.

V. Conclusion

1) PZT-brass composites with redirected stresses exhibit a very high figure of merits ($d_h \cdot g_h$ or $d_h \cdot g_h/\tan \delta$) as well as high dielectric constant K, and high withstanding pressure P_m. The composite characteristics includes values of $d_h \cdot g_h \cong 50{,}000 \times 10^{-15}$ m^2/N, $K >$ 1400 and $P_m > 1000$ psi. The improved transducer performance promises to be important in many naval applications, and for detectors for oil exploration and earthquake seismology.

2) Larger cavity sizes lead to large d_h and g_h, but lower operating frequencies.

3) Very little aging was observed under high hydrostatic pressure of 350 psi.

4) An experimental four-element flexible array shows that the lowest resonance frequency is higher than 30 kHz.

Acknowledgment

The authors wish to thank Dr. W. Smith and Professor L. E. Cross for their advice, and P. A. Marlowe for his help with sample preparation.

References

[1] R. E. Newnham, "Composite electroceramics," *Ann. Rev. Mat. Sci.*, pp. 47–68, Annual Reviews, Inc. (1986).

[2] T. R. Gururaja, A. Safari, R. E. Newnham, and L. E. Cross, "Piezoelectric ceramic-polymer composites for transducer application," *Electronic Ceramics*, L. Levinson, Ed. New York: Marcel Dekker, 1987, pp. 92–128.

[3] H. Banno, "Recent developments of piezoelectric ceramic products and composite of synthetic rubber and piezoelectric ceramic particles," *Ferroelectrics*, vol. 50, no. 1–4, pp. 329–338, 1983.

[4] Y. Q. Zhuang, Y. G. Heu, and Q. C. Xu, "Sandwich PZT/polymer composite transducer," *Ferroelec.*, vol. 49, pp. 241–249, 1983.

[5] G. A. Brigham and L. H. Royster, "Present status in the design of flextensional underwater acoustic transducer," *J. Acoust. Soc. Am.*, vol. 46, no. 1, Pt. 1, p. 92, 1969.

[6] K. D. Rolt, "History of the flextensional electro-acoustic transducer," *J. Acoust. Soc. Am.*, vol. 87, no. 3, pp. 1340–1349, 1990.

[7] R. A. Nelson, Jr. and L. H. Royster, "Development of a mathematical model for the class V flextensional underwater acoustic transducer," *J. Acoust. Soc. Am.*, vol. 49, pp. 1609–1620, 1971.

[8] A. S. Bhalla and R. Y. Ting, "Hydrophone figure of merit," *Sensors and Materials*, vol. 4, pp. 181–185, 1988.

[9] K. L. Swain, "Crescent-shaped 3-0 piezoelectric transducers," B.S. thesis, Dept. Materials Sci. Eng., Pennsylvania State Univ., 1988.

[10] T. J. Meyer, "3-0 crescent shaped composite hydrophones," B.S. thesis, Dept. Materials Sci. Eng., Pennsylvania State Univ., 1989.

Qi Chang Xu was born in Shanghai, China, November 11, 1936.

He is a Senior Research Associate at the Materials Research Laboratory, Pennsylvania State University, after having been a Visiting Associate Professor from 1986 to 1989. His current research interests are in acoustics and ferroelectric materials.

Shoko Yoshikawa was born in Nagao, Japan, on March 25, 1956. She received the B.S. degree in industrial chemistry from the Chubu Institute of Technology, Japan, and the M.S. degree in chemistry in 1981 from Ohio University, Athens, OH.

She has been with the Materials Research Laboratory at Pennsylvania State University, University Park, PA, where she is currently is a Senior Research Assistant working on composite electronic materials. Formerly, she was Senior Chemist working on magnetic tape formulation at Ampex Corp., Magnetic Tape Division R&D, Redwood City, CA, from 1981 to 1984. She also worked on ceramic capacitor formulation at Murata-Erie North America Co., Ltd., and at Centre Engineering, Inc.

John Belsick was born in Rochester, PA, on August 20, 1964. He received the B.S. in ceramic science and engineering in 1986 and the M.S. degree in solid-state science in 1988, both from Pennsylvania State University, University Park, PA, where he is currently pursuing the Ph.D. degree.

In 1989, he was a Visiting Scientist at the Toshiba Corp., Kawasaki, Japan, where he was involved in the development of multilayer capacitors, his research interests are in the areas of ferroelectric thin films, dielectric and piezoelectric ceramics, and composites.

Robert E. Newnham was born in Amsterdam, NY, on March 28, 1929. He received the B.S. degree in mathematics in 1950 from Hartwick College, Oneonta, NY, the M.S. degree in physics and mineralogy, from Colorado State University, Fort Collins, and the Ph.D. in crystallography in 1960 from Cambridge University, Cambridge, England.

He is ALCOA Professor of Solid-State Science at the Materials Research Laboratory at Pennsylvania State University, University Park, PA. Previously, he was a staff member of the Laboratory for Insulation Research at the Massachusetts Institute of Technology, Cambridge, MA. His research interests are in structure-property relations, electroceramics, and composite materials for electronic applications.